

Interactive comment on “Intercomparison of an Aerosol Chemical Speciation Monitor (ACSM) with ambient fine aerosol measurements in Downtown Atlanta, Georgia” by S. H. Budisulistiorini et al.

Anonymous Referee #1

Received and published: 13 February 2014

In this manuscript Budisulistiorini et al. investigate the performance of an ACSM during a long-term deployment in Atlanta, GA by comparing it to various established on-line and offline measurement techniques. Although the ACSM is yet around for several years, literature evaluating its performance is still relatively sparse making this manuscript a valuable addition. The manuscript is well structured and written apart from a few missing articles. However, there are very few significant new findings and some of the conclusions could have been investigated more closely. Altogether I second the publication in AMT after the following comments have been addressed.

General comments:

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- For me it is not identifiable from the manuscript if the correct regression model was used. According to the text, slopes and correlations were calculated using the linear regression model. If this means the ordinary linear regression, it is not sufficient for the case shown. In ordinary linear regression, the sum of the squared vertical distances between the y data values and the corresponding y values on the fitted line are minimized, i.e. only the error of the y values is accounted for. In many cases this can yield different (inverse) slopes depending on which dataset is plotted on the y axis. In a comparison of data from two instruments both axes are error-prone, which demands an orthogonal regression. Only the slopes are affected by this, the coefficients of determination remain valid and the same. Please clarify and, if applicable, refit the data.

- More details about the calibrations of both ACSM would be desirable. Were they calibrated on site using the same equipment for both ACSM? Was the calibration system checked for consistency? How often were the calibrations repeated and how much did the RIEs and RFs vary? Were the same calibration values used for all seasons (obviously not for the SO₄ RIE)? An estimate of the uncertainties of the calibration system would be useful since these of course also propagate into the data.

- The author suggests to not only calibrate the ACSM for NH₄ and NO₃ but also for SO₄ and organics. I see that a calibration of SO₄ was performed and deemed useful, but what about the calibration with organics? Why was this not done? If the authors could suggest a set of organic compounds for such a calibration or even show an example and give values for the organic RIE in the ACSM my concerns about the novelty would be greatly reduced.

- Another point which would add something new to the manuscript is an extension of the comparison of the two ACSM from only the concentrations and time series (section 3.1) to mass spectra. How similar are the mass spectra of two ACSMs sampling the same aerosol, i.e. are there observable differences in the organics fragmentation?

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- The author assumes that chloride was mostly below the detection limit. Were the detection limits of both ACSM measured or estimated?

- An overview about the measured mass loading could be mentioned earlier in the text.

- Suggestions for additional references: AMS: Jayne, J.T., D.C. Leard, X. Zhang, P. Davidovits, K.A. Smith, C.E. Kolb, and D.R. Worsnop, Development of an Aerosol Mass Spectrometer for Size and Composition. *Analysis of Submicron Particles, Aerosol Science and Technology*, 33, 49-70, 2000 CE: A.M. Middlebrook, R. Bahreini, J.L. Jimenez, and M.R. Canagaratna. Evaluation of Composition-Dependent Collection Efficiencies for the Aerodyne Aerosol Mass Spectrometer using Field Data. *Aerosol Science and Technology*, DOI:10.1080/02786826.2011.620041, 46, 258–271, 2012. ACSM intercomparisons: Fröhlich, R., Cubison, M. J., Slowik, J. G., Bukowiecki, N., Prévôt, A. S. H., Baltensperger, U., Schneider, J., Kimmel, J. R., Gonin, M., Rohner, U., Worsnop, D. R., and Jayne, J. T.: The ToF-ACSM: a portable aerosol chemical speciation monitor with TOFMS detection, *Atmos. Meas. Tech.*, 6, 3225-3241, doi:10.5194/amt-6-3225-2013, 2013.

- I have doubts about the presented evidence for organic nitrates at m/z 30. The authors plot m/z 30 and m/z 46 against the JST NO₃ and conclude that the higher signals at m/z 30 in fall suggest that there are additional signals other than NO₃ in the m/z 30 channel. For me the situation in fall (Fig. S9(b)) looks more to come from a non-constant nitrate fragmentation which depends on the total nitrate loading. This was a known ACSM issue, originating from the internal detector settings and are resolved since DAQ version 1.4.3.8. It seems as if the m/z 30 decreases at higher loadings while the m/z 46 increases. If this was the case in the examined dataset can be found out by plotting m/z 46 vs m/z 30 and checking for a non-linearity there. In Fig. S8(a) these trends do not show because of the lower loadings. I suggest to re-check the fits in fall after excluding all points higher than approx. 1 $\mu\text{g}/\text{m}^3$ to look at about the same concentration range as in summer and see if the observed slope difference can still be observed.

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- Regressions in suppl. Fig. S10 and S11: since the distributions are heteroskedastic one of the assumptions of the classical regression model is violated and one has to take special care about the confidence intervals. Coefficients of determination and slopes should still be valid, but the given confidence intervals are blurred and likely incorrect.

Specific comments:

The referee is a non-native English speaker. Hence, language comments are merely suggestions.

- “Collocated” or “co-located”? (e.g. P11183 L10)

- P11184 L5: Close bracket after “. . .OC”

- P11184 L27: the instead of their

- P11186 L7: replace “as from” with “like”. Here the authors should add that the ACSM not only has reduced sensitivity but also lower mass resolution than the AMS. Numbers about sensitivity and mass resolution would also be useful

- P11186 L9: “lower grade quadrupole”. Lower than what? Please specify.

- P11188 L7: “amu⁽⁻¹⁾” could be misunderstood as “amu*s⁽⁻¹⁾”

- P11189 L9: The range of SO₄ RIEs used is relatively large (0.44-1.10). Does this represent the uncertainty of the fitting approach or is there a seasonal variation?

- P11189 L23: which assumptions?

- P11191 L3ff: At first look this seems to be circular, the continuous measurements are adjusted to the filters and then correlate well with the filters. The names “Level_1” and “Level_2” are obsolete, they are only used at one more instance in the manuscript.

- P11193 L2: The authors supposedly do not mean to find meteorological variations across seasons as stated in the text but variations of aerosol parameters

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- P11193 L3-L8: I suggest dropping this paragraph. Repetition of content from Sect. 3.2.1
- P11194 L3: Please recheck the slope of 0.67 from the summer intercomparison of NH4. After having a look at Fig. S6(d) it seems too low.
- P11194 L7: Please clarify the baseline comment. I am not aware of a baseline calculation in the ACSM software.
- P11194 L8: "slope" instead of "slopes"
- P11195 L4: how is the +/-47% variation calculated?
- P11195 L8: please specify meteorological influence
- P11197 L22: Where does this 30% uncertainty estimation come from?
- Sect. 4.3: I suggest to include information about the "neutralization balance (NH4 measured vs NH4 predicted)" for both ACSM and continuous measurements
- P11200 L24: remove "in". An incorrect RIE of organics would produce a constant offset, this could be pointed out more precisely.
- P11201 L10: why is this the "traditional ambient aerosol density"? The cited 1.2 g cm^{-3} is an organic aerosol density according to Turpin et al. Published overall densities of ambient aerosol are usually higher. E.g. a similar density of 1.61 g cm^{-3} was reported from Beijing. Cf.: Hu, Min, et al. "Estimation of Size-Resolved Ambient Particle Density Based on the Measurement of Aerosol Number, Mass, and Chemical Size Distributions in the Winter in Beijing." *Environmental Science & Technology* 46.18 (2012): 9941-9947.
- P11201 L13: This last sentence is very general.
- P11202 L5: The AMS is not really the predecessor of the ACSM. The instruments are related, but both have different fields of application and reasons of existence.

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- Missing authors in the following citations: Jimenez 2009, Peltier 2007, Polidoro 2008, Schwab 2003, Takegawa 2005 and Turpin 2001
- Suppl P4 L2: The standard RIE of SO4 which should be used for the fitting is 1.2 not 1.5, is it?
- Suppl Fig. S4 (a): Do the authors have a hypothesis why the sunset OC seems to have such a large offset?
- Suppl Fig. S7 and S6: In my opinion these figures could also go into the main document.

Interactive comment on Atmos. Meas. Tech. Discuss., 6, 11181, 2013.

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